A sufficient condition for partial ensemble controllability of bilinear Schrödinger equations with bounded coupling terms

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A Sufficient Condition for Partial Ensemble Controllability of Bilinear Schrödinger Equations with Bounded Coupling Terms

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***Abstract*— This note presents a sufficient condition for partial approximate ensemble controllability of a set of bilinear conservative systems in an infinite dimensional Hilbert space. The proof relies on classical geometric and averaging control techniques applied on finite dimensional approximation of the infinite dimensional system. The results are illustrated with the planar rotation of a linear molecule.**

I. INTRODUCTION

A. Control of quantum systems

The state of a quantum system evolving in a Riemannian manifold Ω is described by its *wave function*, a point ψ in $L^2(\Omega, \mathbf{C})$. When the system is submitted to an electric field (e.g., a laser), the time evolution of the wave function is given, under the dipolar approximation and neglecting decoherence, by the Schrödinger bilinear equation:

$$\mathrm{i}\frac{\partial \psi}{\partial t} = (-\Delta + V(x))\psi(x,t) + u(t)W(x)\psi(x,t) \qquad (1)$$

where Δ is the Laplace-Beltrami operator on Ω, V and W are real potential accounting for the properties of the free system and the control field respectively, while the real function of the time u accounts for the intensity of the laser.

In view of applications (for instance in NMR), it is important to know whether and how it is possible to chose a suitable control $u : [0,T] \to \mathbf{R}$ in order to steer (1) from a given initial state to a given target. This question has raised considerable interest in the community in the last decade. After the negative results of [1] and [2] excluding exact controllability on the natural domain of the operator $-\Delta + V$ when W is bounded, the first, and at this day the only one, description of the attainable set for an example of bilinear quantum system was obtained by Beauchard ([3], [4]). Further investigations of the approximate controllability of (1) were conducted using Lyapunov techniques ([5], [6], [7], [8], [9], [10]) and geometric techniques ([11], [12]).

B. Ensemble controllability

In many applications, a macroscopic device acts on a large number of identical microscopic quantum systems (for instance, a single laser acts on a small quantity of liquid containing many molecules). Usually, the external field acts differently on each of the small systems (depending for instance on the orientation of the molecule with respect to the exterior electric field). For the sake of simplicity, we will assume in this work that the action of the external field on the system a is proportional to a. Instead of one system of type (1), one has to control a continuum:

$$\mathrm{i}\frac{\partial \psi_\alpha}{\partial t} = (-\Delta + V(x) + u(t)\alpha W(x))\psi_\alpha(x,t) \qquad (2)$$

where the system labeled with α, $\alpha \in [0,1]$, has wave function ψ_α. Notice that, since the systems are physically identical, the free dynamics (when $u=0$) is the same for every a.

The simultaneous (or *ensemble*) control problem turns into the following question: let a continuum of initial conditions $(\psi^0_a)_{a\in[0,1]}$ and of targets $(\psi^t_a)_{a\in[0,1]}$ be given. Does it exist a control u that steers the systems (2) from ψ^0_a to ψ^t_a for every a in $[0,1]$?

Because of its crucial importance for applications (dispersion of parameters is *always* present in real world systems), the problem of *ensemble controllability* of quantum systems has been tackled by many authors, see for instance [13], [14], [15], [16], [17], [18] for theoretical results and [19] for numerical aspects.

C. Framework and notations

To take advantage of the powerful tools of linear operators, we will reformulate the problem (2) in the following abstract setting. Let H be a separable Hilbert space, endowed with the $\langle\cdot,\cdot\rangle$ Hilbert product. We consider the continuum of control systems

$$\frac{\mathrm{d}\psi}{\mathrm{d}t} = A\psi + u(t)\alpha B\psi, \quad \alpha \in [0,1], \qquad (3)$$

where the linear operators A, B can be completed in a 5- or 6-uple that satisfies Assumption 1 or Assumption 2.

Assumption 1: The 6-uple $(A, B, \lambda_1, \phi_1, \lambda_2, \phi_2)$ satisfies

1) A is skew-adjoint with domain $D(A)$;
2) B is bounded and skew-symmetric;
3) ϕ_1 and ϕ_2 are two eigenvectors of A of norm 1, associated with eigenvalues $-\mathrm{i}\lambda_1$ and $-\mathrm{i}\lambda_2$;
4) $\lambda_1 < \lambda_2$ and $\langle \phi_1, B\phi_2\rangle \neq 0$;
5) for every eigenvalues $-\mathrm{i}\mu$ and $-\mathrm{i}\mu'$ of A, associated with eigenvectors v and v', $|\lambda_1 - \lambda_2| = |\mu - \mu'|$ implies $\{\lambda_1,\lambda_2\} = \{\mu,\mu'\}$ or $\{\lambda_1,\lambda_2\} \cap \{\mu,\mu'\} = \emptyset$ or $\langle v, Bv'\rangle = 0$;

6) the essential spectrum of $\mathrm{i}A$ (if any) does not accumulate in any of these four points: $2\lambda_1 - \lambda_2, \lambda_1, \lambda_2$ and $2\lambda_2 - \lambda_1$.

Assumption 2: The 5-uple (A, B, U, Λ, Φ) satisfies

1) A is skew-adjoint with domain $D(A)$;
2) B is skew-symmetric;
3) U is a subset of $\mathbf{R}$ containing at least 0 and the points $\{1/n, n \in \mathbf{N}\}$;
4) for every u in U, $A+uB$ is skew-adjoint (with domain not necessarily equal to $D(A)$);
5) $\Phi = (\phi_j)_{j\in\mathbf{N}}$ is a Hilbert basis of H made of eigenvectors of A, all of which in the domain of B;
6) $\Lambda = (\lambda_j)_{j\in\mathbf{N}}$ is a sequence of real numbers such that, for every j in $\mathbf{N}$, $A\phi_j = -\mathrm{i}\lambda_j$;
7) $\lambda_1 < \lambda_2$ and $\langle \phi_1, B\phi_2\rangle \neq 0$;
8) for all eigenvalues $-\mathrm{i}\mu$ and $-\mathrm{i}\mu'$ of A, associated with eigenvectors v and v', $|\lambda_1 - \lambda_2| = |\mu - \mu'|$ implies $\{\lambda_1, \lambda_2\} = \{\mu, \mu'\}$ or $\{\lambda_1, \lambda_2\} \cap \{\mu, \mu'\} = \emptyset$ or $\langle v, Bv'\rangle = 0$.

If (A, B) satisfies Assumptions 1.1 and 1.2 (resp. (A, B, U) satisfies Assumptions 2.1, 2.3 and 2.4), then for every t_0, t in $\mathbf{R}$, for every u in $L^1(\mathbf{R}, \mathbf{R})$ (resp. $u : \mathbf{R} \to U$ piecewise constant), there exists a unique family of unitary operators $(\Upsilon^{u,\alpha}_{t,t_0})_{\alpha\in[0,1]}$ such that, for every family $(\psi^0_\alpha)_{\alpha\in[0,1]}$ in H, for every α in $[0, 1]$, $t \mapsto \Upsilon^{u,\alpha}_{t,t_0}\psi^0_\alpha$ is the unique solution of (3) in the weak sense that satisfies $\Upsilon^{u,\alpha}_{t_0,t_0}\psi^0_\alpha = \psi^0_\alpha$.

Let $(A, B, \lambda_1, \phi_1, \lambda_2, \phi_2)$ satisfy Assumption 1 or (A, B, U, Λ, ϕ) satisfy Assumption 2. We define the 2-dimensional Hilbert space $\mathcal{L}_2 = \mathrm{span}(\phi_1, \phi_2)$, $\pi_2 : H \to H$ the orthogonal projection on $\mathcal{L}_2$, $A^{(2)} = \pi_2 A\pi_2$ and $B^{(2)} = \pi_2 A\pi_2$ the compressions of A and B on $\mathcal{L}_2$, and $X^{u,\alpha}_{(2)}$, the propagator associated with the (infinite dimensional) system $x' = A^{(2)}x + u\alpha B^{(2)}x$. By abuse, we will still denote with $X^{u,\alpha}_{(2)}$ the restriction of $X^{u,\alpha}_{(2)}$ to $\mathcal{L}_2$.

D. Main result

Proposition 1: Let $(A, B, \lambda_1, \phi_1, \lambda_2, \phi_2)$ satisfy Assumption 1 (resp. (A, B, U, Λ, Φ) satisfy Assumption 2) and let $\hat{\Upsilon} : \alpha \in [0, 1] \mapsto \hat{\Upsilon}^\alpha \in U(\mathcal{L}_2)$ be a continuous curve of unitary operators on $\mathcal{L}_2$ that satisfies $\hat{\Upsilon}^0 = \mathrm{Id}_{\mathcal{L}_2}$. Then, for every $\varepsilon > 0$, for every $\delta > 0$, there exists $u : [0, T] \to [-\delta, \delta]$ (resp. piecewise constant with value in $U \cap [0, \delta]$) such that for every α in $[0, 1]$ and (j, k) in $\{1, 2\} \times \{1, 2\}$, $\left| |\langle \phi_j, \Upsilon^{u,\alpha}_{T,0}\phi_k\rangle| - |\langle \phi_j, \hat{\Upsilon}^\alpha\phi_k\rangle| \right| < \varepsilon$.

In other words, up to an arbitrary small error ε, it is possible to steer the eigenvectors ϕ_1 and ϕ_2, simultaneously for every α, to a target with prescribed modulus of coordinates on ϕ_1 and ϕ_2.

The contribution of this note relies on the very same idea as [13], namely the computation of finite dimensional Lie brackets and a polynomial interpolation. The only novelty is that all the steps of the proof come along with explicit estimates, which allow to let the dimension of the finite dimensional systems tend to infinity and eventually prove the infinite dimensional result.

The main improvements of this work with respect to the cited references are

- the possibly infinite dimension of the ambient space H;
- the possibility for the spectrum of A to have a continuous part;
- the possible (finite or not) degeneracy (or multiplicity) of the eigenvalues of A;
- (in the case of Assumption 2) the possible unboundedness of operator B with respect to A, that is, in a case where Kato-Rellich theorem does not apply to $A+uB$.

E. Content of the paper

The core of the proof of Proposition 1 is a constructive approximate controllability result about the propagator $X^{u,\alpha}_{(n)}$ in some finite dimensional subspaces $\mathcal{L}_n$ of H proved in Section II. The precise estimates of Section II-C allows to let the dimension of $\mathcal{L}_n$ tend to infinity and eventually to prove, in Section III-B, the infinite dimensional result for systems satisfying Assumption 2. In Section III-C, we will see that the convergence process used for the proof of section III-B is actually robust enough with respect to perturbation of the spectrum of A to ensure convergence also for systems satisfying Assumption 1. The results are applied to the example of the 3D rotation of a collection of linear molecules in Section IV.

II. FINITE DIMENSIONAL PRELIMINARIES

A. Notations and result

Let N in $\mathbf{N}$, $A^{(N)}, B^{(N)}$ be two matrices in $\mathfrak{u}(N)$ (that is, $\overline{A^{(N)}}^T + A^{(N)} = \overline{B^{(N)}}^T + B^{(N)} = 0$). We consider the continuum of N-dimensional systems

$$x' = A^{(N)} + u\alpha B^{(N)}x, \quad \alpha \in [0, 1] \tag{4}$$

where x is a point in $\mathbf{C}^N$ endowed with its canonical Hilbert structure $\langle\cdot,\cdot,\rangle$. For every locally integrable function u, we define $X^{u,\alpha}_{(N)}$ the propagator associated with (4).

We assume that $A^{(N)}$ is diagonal in $(\phi_j)_{j\leq N}$, the canonical basis of $\mathbf{C}^N$, we denote with $(-\mathrm{i}\lambda_j)_{j\leq N}$ the diagonal of $A^{(N)}$ and with $b_{jk} := \langle \phi_j, B^{(N)}\phi_k\rangle, 1 \leq j, k \leq N$ the entries of $B^{(N)}$. For every $j \leq N$, we define π_j, the orthogonal projection of $\mathbf{C}^N$ to $\mathcal{L}_j = \mathrm{span}(\phi_1, \ldots, \phi_j)$.

Proposition 2: Assume that $(A^{(N)}, B^{(N)}, \lambda_1, \phi_1, \lambda_2, \phi_2)$ satisfies Assumption 1. Let $\hat{\Upsilon} : \alpha \in [0, 1] \mapsto \hat{\Upsilon}^\alpha \in U(\mathcal{L}_2)$ be a continuous curve of unitary operators on $\mathcal{L}_2$ that satisfies $\hat{\Upsilon}^0 = \mathrm{Id}_{\mathcal{L}_2}$. Then, for every $\varepsilon > 0$, for every $\delta > 0$, there exists $u : [0, T] \to [-\delta, \delta]$ such that, for (j, k) in $\{1, 2\}^2$, $\left| |\langle \phi_j, \Upsilon^{u,\alpha}_{T,0}\phi_k\rangle| - |\langle \phi_j, \hat{\Upsilon}^\alpha\phi_k\rangle| \right| < \varepsilon$.

The proof of Proposition 2 is split in two steps. In a first time, after a suitable change of variable, we introduce a continuum of two-dimensional auxiliary systems in Section II-B. Classical Lie groups technique, and the associated uniform convergence estimates, to prove approximate ensemble controllability of these systems. In a second time, in Section II-C, we use classical averaging techniques to show that the trajectories of the systems introduced in Section II-B can be tracked, with arbitrary precision, by the system (4).

B. An auxiliary system

We consider the continuum of control systems in $\mathbf{U}(2)$

$$x'_\alpha = \alpha \begin{pmatrix} b_{11} & b_{12}e^{\mathrm{i}\theta} \\ b_{21}e^{-\mathrm{i}\theta} & b_{22} \end{pmatrix} x_\alpha, \quad \alpha \in [0,1] \tag{5}$$

with initial condition $x_\alpha(0) = I_2$ and control function $\theta : \mathbf{R} \to \mathbf{R}$. For every piecewise constant function $\theta : \mathbf{R} \to \mathbf{R}$, for every α, we denote with $Y^{\theta,\alpha}$ the propagator of (5).

Proposition 3: Let $\hat{\Upsilon} : \alpha \in [0,1] \mapsto \hat{\Upsilon}^\alpha \in SU(\mathcal{L}_2)$ be a continuous curve of unitary operators on $\mathcal{L}_2$ that satisfies $\hat{\Upsilon}^0 = \mathrm{Id}_{\mathcal{L}_2}$. Then, for every $\varepsilon > 0$, there exists $u : [0,T] \to [-\pi,\pi]$ piecewise constant such that, for (j,k) in $\{1,2\}^2$,

$$\left| |\langle \phi_j, Y_T^{\theta,\alpha}\phi_k\rangle| - |\langle \phi_j, \hat{\Upsilon}^\alpha \phi_k\rangle| \right| < \varepsilon.$$

Proof: Let $\varepsilon > 0$. There exists a continuous function $\tilde{v} : \alpha \mapsto \tilde{v}^\alpha \in \mathfrak{su}(\mathcal{L}_2)$ such that, for every α in $[0,1]$, $\|\exp(\tilde{v}^\alpha) - \hat{\Upsilon}^\alpha\| < \varepsilon$. By density of odd polynomials mapping, for the norm of uniform convergence, in the set of odd continuous functions, there exists a polynomial mapping $P : \alpha \mapsto P_\alpha = \sum_{l=0}^N \alpha^{2l+1} Z_{2l+1}$, with $Z_1, Z_3, \ldots, Z_{2N+1}$ in $\mathfrak{su}(2)$ such that $\|P_\alpha - \tilde{v}^\alpha\| < \varepsilon$ for every α in $[0,1]$.

Lemma 4: Let X and Y two matrices in $\mathfrak{su}(2)$, and $(C_j(X,Y))_{1\leq j\leq p}$ a sequence of iterated brackets of X and Y. We denote with l_j the length of the bracket $C_j(X,Y)$ (the length of $[X,Y]$ is 1). Then, for every real sequence $(\beta_j)_{1\leq j \leq p}$, for every T in $\mathbf{R}$, for every $\varepsilon > 0$, there exists a finite sequence $(t_k)_{1\leq k\leq m}$ in $\mathbf{R}$ such that, for every α in $[0,1]$, $\|P_\alpha - e^{T\sum_{j=1}^p \beta_j \alpha^{l_j} C_j(X,Y)}\| < \varepsilon$, where P_α is the product of matrices $e^{t_1\alpha X}e^{t_2\alpha Y}\cdots e^{t_{m-1}\alpha X}e^{t_m \alpha Y}$.

Proof: This result is very classical when $\alpha = 1$ (i.e., one considers one system only). The uniform version presented here (with α in $[0,1]$) is basically contained in [17]. Because of its importance for our purpose, we give below a sketch of the proof of the result.

We first assume that $p = 1$ and we proceed by induction on the length of C_1. From the Baker-Campbell-Hausdorff formula, we deduce that, for every 2×2 matrices X, Y, there exists a function $g_{X,Y} : \mathbf{R} \to \mathfrak{gl}_2$ tending to $0_{2\times 2}$ at 0 such that, for every t in $\mathbf{R}$, for every α in $[0,1]$,

$$e^{t\alpha X}e^{t\alpha Y}e^{-t\alpha X}e^{-t\alpha Y} = e^{\alpha^2 t^2 [X,Y] + g_{X,Y}(\alpha t)\alpha^2 t^2}.$$

As a consequence, for every 2×2 matrices X and Y,

$$\lim_{t\to 0} \frac{1}{t^2} \left\| e^{t\alpha X}e^{t\alpha Y}e^{-t\alpha X}e^{-t\alpha Y} - e^{\alpha^2 t^2 [X,Y]} \right\| = 0, \tag{6}$$

the convergence being uniform with respect to α in $[0,1]$.

Recall that, for every V, W in $\mathfrak{su}(2)$, for every n in $\mathbf{N}$,

$$\begin{aligned} \|V^n - W^n\| &= V(V^{n-1} - W^{n-1}) + (V-W)W^{n-1} \\ &\leq \|V^{n-1} - W^{n-1}\| + \|V - W\| \\ &\leq n\|V - W\|. \end{aligned} \tag{7}$$

Hence,

$$\left\| \left(e^{t\alpha X}e^{t\alpha Y}e^{-t\alpha X}e^{-t\alpha Y}\right)^n - e^{n\alpha^2 t^2[X,Y]} \right\| \leq n \left\| e^{t\alpha X}e^{t\alpha Y}e^{-t\alpha X}e^{-t\alpha Y} - e^{\alpha^2 t^2 [X,Y]}\right\| \tag{8}$$

Choosing $n = T/t^2$ and letting n tend to infinity (and hence t tend to zero) gives the result for $p = 1$, $\beta_1 = 1$ and $l_1 = 1$. The proof for $l_1 > 1$ is very similar, replacing X and Y by the suitable iterated brackets in (8).

A consequence of Zassenhauss formula is that, for every 2×2 matrices U, V, there exists a locally Lipschitz function $g : \mathfrak{gl}(2) \times \mathfrak{gl}(2) \times \mathbf{R} \to \mathbf{R}$ that vanishes as soon as one of its entries vanishes such that, for every t in $\mathbf{R}$, for every α in $[0,1]$, for every j,k in $\mathbf{N}$,

$$\|e^{tU}e^{tV} - e^{t(U+V)}\| \leq t^2 g(U,V,t).$$

The proof of Lemma 4, for $p > 1$ and β_j not necessarily equal to 1, follows by choosing $t = T/n$ for n large enough and using once again (7). ■

We come back to the proof of Proposition 3. After the time dependent change of variable

$$y_\alpha = \exp\left[-t\alpha \begin{pmatrix} b_{11} & 0 \\ 0 & b_{22}\end{pmatrix}\right] x_\alpha, \tag{9}$$

the system (5) reads

$$y'_\alpha = \alpha \begin{pmatrix} 0 & b_{12}e^{\mathrm{i}\theta - t(b_{11}-b_{22})} \\ b_{21}e^{-\mathrm{i}\theta + t(b_{11}-b_{22})} & 0 \end{pmatrix} y_\alpha, \tag{10}$$

or $y'_\alpha = M_\alpha^\nu y_\alpha$, defining $\nu = \theta - \mathrm{i}t(b_{11} - b_{22})$, with

$$M_\alpha^\nu = \alpha \begin{pmatrix} 0 & b_{12}e^{\mathrm{i}\nu} \\ b_{21}e^{-\mathrm{i}\nu} & 0 \end{pmatrix}. \tag{11}$$

Lemma 5: For every ϕ in $\mathbf{C}^2$, for every t in $\mathbf{R}$, for every locally integrable $\theta : \mathbf{R} \to \mathbf{R}$, the moduli of the coordinates in the canonical basis (ϕ_1, ϕ_2) of $\mathbf{C}^2$ of $y_\alpha(t)\psi$ and $x_\alpha(t)\psi$ are the same.

Proof: From (9), the coordinates of y_α and x_α are equal, up to a phase shift depending on time and α. ■

Thanks to Lemma 4, Proposition 3 follows if, for every l, the matrix Z_{2l+1} defined above can be realized as a linear combination (with real coeeficients) of brackets of length exactly equal to $2l+1$ of the matrices $M_1^\nu, \nu \in [-\pi,\pi]$. Notice that

$$M_1^0 = \begin{pmatrix} 0 & b_{12} \\ b_{21} & 0 \end{pmatrix} \text{ and } M_1^{\frac{\pi}{2}} = 1 \begin{pmatrix} 0 & \mathrm{i}b_{12} \\ -\mathrm{i}b_{21} & 0\end{pmatrix}.$$

Straightforward computations give, for every k in $\mathbf{N}$,

$$ad^k_{M_\alpha^{\frac{\pi}{2}}} M_\alpha^0 = \alpha^{2k+1} \begin{pmatrix} 0 & b_{12}|b_{12}|^{2k} \\ b_{21}|b_{12}|^{2k} & 0 \end{pmatrix}, \tag{12}$$

Proposition 3 follows from the fact that $b_{12} \neq 0$. ■

C. Averaging techniques

We define the $N \times N$ matrix N_α^θ by $N_\alpha^\theta(j,k) = 0$ for every j,k in $\{1,\ldots,N\}^2$ but $N_\alpha^\theta(1,2) = \alpha b_{12}e^{\mathrm{i}\theta}$ and $N_\alpha^\theta(2,,1) = -\overline{N_\alpha^\theta(1,2)}$. In particular, N_α^θ belongs to $\mathfrak{su}(N)$ and $\pi_2 N_\alpha^\theta \pi_2 = M_\alpha^\theta$.

Let us come back to the proof of Proposition 2. From Proposition 3, it is enough to show that, for every θ, t in $\mathbf{R}$ and every $\varepsilon > 0$, there exists $u_\varepsilon : [0, T_\varepsilon] \to (-\delta, \delta)$ such that, for every α in $[0,1]$, $\|\pi_2 X_{(N)}^{u_\varepsilon}(T_\varepsilon) - e^{tN_\alpha^\theta}\| < \varepsilon$. This is exactly the content of Proposition 6, whose proof is given in [20].

Proposition 6: Let $u^* : \mathbf{R}^+ \to \mathbf{R}$ be a locally integrable function.

Assume that u^* is periodic with period $T = \frac{2\pi}{|\lambda_2 - \lambda_1|}$ and that $\int_0^T u^*(\tau)e^{\mathrm{i}(\lambda_l - \lambda_m)\tau}\mathrm{d}\tau = 0$ for every $\{l,m\}$ such that $\{l,m\} \cap \{1,2\} \neq \emptyset$ and $\lambda_l - \lambda_m \in (\mathbf{Z} \setminus \{\pm 1\})(\lambda_1 - \lambda_2)$ and $b_{lm} \neq 0$. For every n, define $v_n : t \mapsto 1/n \int_0^t |u^*(s)|\mathrm{d}s$ and the $N \times N$ matrix $M^\dagger$ with entries $m^\dagger_{j,k} = b_{jk}\int_0^T u^*(s)e^{\mathrm{i}(\lambda_2-\lambda_1)s}\mathrm{d}s / \int_0^T |u^*(s)|\mathrm{d}s$.

If $\int_0^T u^*(\tau)e^{\mathrm{i}(\lambda_2-\lambda_1)\tau}\mathrm{d}\tau \neq 0$, then, for every n in $\mathbf{N}$, for every $t \leq nT^*$,

$$\frac{\|X^{u_n}_{(N)}(t,0) - e^{tA^{(N)}}e^{v_n^{[-1]}(t)M^\dagger}\|}{I(C+1)\|B^{(N)}\|} \leq \frac{1 + 2K\|B^{(N)}\|}{n}. \quad (13)$$

with

$$T^* = \frac{\pi T}{2|b_{1,2}|\left|\int_0^T u^*(\tau)e^{\mathrm{i}(\lambda_1-\lambda_2)\tau}\mathrm{d}\tau\right|}, \quad I = \int_0^T |u^*(\tau)|\mathrm{d}\tau,$$

$$K = \frac{IT^*}{T}, C = \sup_{(j,k)\in\Lambda}\left|\frac{\int_0^T u^*(\tau)e^{\mathrm{i}(\lambda_j-\lambda_k)\tau}\mathrm{d}\tau}{\sin\left(\pi\frac{|\lambda_j-\lambda_k|}{|\lambda_2-\lambda_1|}\right)}\right|,$$

where Λ is the set of all pairs (j,k) in $\{1,\ldots,N\}^2$ such that $b_{jk} \neq 0$ and $\{j,k\} \cap \{1,2\} \neq \emptyset$ and $|\lambda_j - \lambda_k| \notin \mathbf{Z}|\lambda_2 - \lambda_1|$.

Proof: (Proposition 2) We apply Proposition 6 with u^* periodic with period $T = \frac{2\pi}{|\lambda_2-\lambda_1|}$ and satisfying $\int_0^T u^*(\tau)e^{\mathrm{i}(\lambda_2-\lambda_1)\tau}\mathrm{d}\tau \neq 0$ $\int_0^T u^*(\tau)e^{\mathrm{i}(\lambda_l-\lambda_m)\tau}\mathrm{d}\tau = 0$ for every $\{l,m\}$ such that $\{l,m\}\cap\{1,2\} \neq \emptyset$ and $\lambda_l - \lambda_m \in (\mathbf{Z}\setminus\{\pm1\})(\lambda_1 - \lambda_2)$ and $b_{lm} \neq 0$. Such a u^* can be chosen of the form $t \mapsto \cos((\lambda_2 - \lambda_1)t - \theta)$ or piecewise constant with value in $\{0,1\}$ (for an explicit construction of such a function, see [12]).

To ensure that $e^{v_n^{[-1]}(t)M^\dagger}$ is ε-close to $e^{tM^\nu_\alpha}$, defined as in (11), one chooses t such that $v_n(t) = b_{12}e^{\mathrm{i}\theta}/m^\dagger_{12} \leq nT^*$. One can check from the definition of v_n that $t/(nr^*)$ tends to 1 as n tends to infinity, where r^* is defined by

$$r^* = \frac{T}{I}v_1^{[-1]}\left(\frac{b_{12}e^{\mathrm{i}\theta}}{m^\dagger_{12}}\right).$$

The final step in the proof of Proposition 2 is to get rid of the phase $e^{tA^{(N)}}$ in estimate (13). We use the Poincaré Recurrence Theorem with the mapping

$$\begin{array}{ccc} \mathbf{R} & \to & \mathbf{R}^{n+1}/\mathbf{Z}^{n+1} \\ s & \mapsto & \left(\frac{\lambda_1}{2\pi}s, \frac{\lambda_2}{2\pi}s, \ldots, \frac{\lambda_n}{2\pi}s, \frac{1}{r^*}s\right) \end{array}$$

on the $n+1$ dimensional torus. For every $\varepsilon > 0$, there exists a sequence $(s_k)_{k\in\mathbf{N}}$ that tends to infinity such that $s_k\lambda_j$ is ε close to $2\pi\mathbf{Z}$ and s_k is ε-close to $r^*\mathbf{Z}$. The sequence of controls $\lfloor r^*/s_k\rfloor u^*$ gives Proposition 2 by letting k tend to infinity. ■

III. INFINITE DIMENSIONAL ESTIMATES

A. Heuristic of the proof

In this Section, we proceed to the proof of Proposition 1. Inspired by Section II, it is enough to show that the projections of each of the infinite dimensional systems (2) can track, with an arbitrary precision, the trajectories of the 2×2 system (5).

To begin with, we consider in Section III-B a system that satisfies Assumption 2. The proof is a uniform version of the Section 4 of [20] which is valid for one particular α.

To prove Proposition 1 for systems that satisfy Assumption 1, we first estimate the robustness of the results of Section III-B against a perturbation of the spectrum of A. The conclusion will follow from the Von Neumann approximation theorem. As in Section III-B, the method of the proof in Section III-C is similar to the one used in [20], the only difference lying once again in the uniformity of the convergence estimates with respect to α in $[0,1]$.

B. If the eigenvectors of A span a dense subspace of H

Let (A,B,U,Λ,Φ) satisfy Assumption 2, θ in $[-\pi,\pi]$ and $r,\varepsilon > 0$. We aim to find a periodic control u^* with period $2\pi/(\lambda_2 - \lambda_1)$, n in $\mathbf{N}$ and T in $\mathbf{R}^+$ such that, for every α in $[0,1]$,

$$\|\Upsilon^{u^*/n,\alpha}_{T,0} - e^{rM^\theta_\alpha}\| < \varepsilon.$$

Since ϕ_1 and ϕ_2 belong to the domain of B, the sequences $(b_{1,l})_{l\in\mathbf{N}}$ and $(b_{2,l})_{l\in\mathbf{N}}$ are in ℓ^2. Hence, there exists N in $\mathbf{N}$ such that $\|\pi_2 B(1-\pi_N)\| = \|(1-\pi_N)B\pi_2\| < 5\varepsilon/(2r)$. Define $\omega = \lambda_2 - \lambda_1$ and u^* with period $2\pi/\omega$ in such a way that the $N\times N$ matrix $M^\dagger$ of Proposition 6 is equal to M^θ_1 and the efficiency $|\int_0^{\frac{2\pi}{\omega}} u^*(s)e^{\mathrm{i}\omega s}\mathrm{d}s| / \int_0^{\frac{2\pi}{\omega}} |u^*(s)|\mathrm{d}s$ of u^* for the transition $(1,2)$ is larger than $2/5$. This can be done, for instance, with $t \mapsto \cos(wt - \theta)$ (efficiency $\pi/4$) in the case where B is bounded or, in the general case of Assumption 2, with a piecewise constant function taking value in $\{0,1\}$ as described in [12].

For a given n to be precised later, consider system (3) with control $u_n = u^*/n$ in projection on $\mathrm{span}(\phi_1,\ldots,\phi_N)$:

$$\begin{aligned}\pi_N\frac{\mathrm{d}}{\mathrm{d}t}\Upsilon^{u_n,\alpha}_t\phi_j &= (A^{(N)} + u_n(t)\alpha B^{(N)})\Upsilon^{u_n,\alpha}_t\phi_j \\ &\quad + u_n(t)\pi_N\alpha B(1-\pi_N)\Upsilon^{u_n,\alpha}_t\phi_j. \quad (14)\end{aligned}$$

From the variation of the constant, we get, for $j = 1,2$,

$$\begin{aligned}\pi_N\Upsilon^{u_n,\alpha}_t\phi_j &= X^{u_n,\alpha}_{(N)}(t,0)\phi_j \\ &+ \int_0^t u_n(s)X^{u_n,\alpha}_{(N)}(t,s)\pi_N\alpha B(1-\pi_N)\Upsilon^{u_n,\alpha}_t\phi_j\mathrm{d}s. \quad (15)\end{aligned}$$

Project (15) on $\mathrm{span}(\phi_1,\phi_2)$, and recall that $\pi_N\pi_2 = \pi_2\pi_N = \pi_2$ for $N \geq 2$:

$$\begin{aligned}\pi_2\Upsilon^{u_n,\alpha}_t\phi_j &= \pi_2 X^{u_n,\alpha}_{(N)}(t,0)\phi_j \quad (16) \\ &+ \int_0^t u_n(s)\pi_2 X^{u_n,\alpha}_{(N)}(t,s)\pi_N\alpha B(1-\pi_N)\Upsilon^{u_n,\alpha}_t\phi_j\mathrm{d}s.\end{aligned}$$

Define, for every t, s in $\mathbf{R}$, the bounded linear mapping $[\pi_2, X^{u_n,\alpha}_{(N)}(t,s)] := \pi_2 \circ X^{u_n,\alpha}_{(N)}(t,s) - X^{u_n,\alpha}_{(N)}(t,s) \circ \pi_2$. Equation (16) reads, for $j = 1, 2$,

$$\begin{aligned}
&\pi_2 \Upsilon^{u_n,\alpha}_t \phi_j - \pi_2 X^{u_n,\alpha}_{(N)}(t,0)\phi_j = \\
&\quad -\int_0^t u_n(s) X^{u_n,\alpha}_{(N)}(t,s)\pi_2 \alpha B(1-\pi_N)\Upsilon^{u_n,\alpha}_t \phi_j \mathrm{d}s \quad (17)\\
&\quad +\int_0^t u_n(s)[\pi_2, X^{u_n,\alpha}_{(N)}(t,s)]\pi_N \alpha B(1-\pi_N)\Upsilon^{u_n,\alpha}_t \phi_j \mathrm{d}s.
\end{aligned}$$

Extend the definition of M^α_θ to H by $M^\dagger = 0$ on $\mathcal{L}_N^\perp$ and define the linear operator $E^{n,\alpha}_N(t) := X^{u_n}_{(N)}(t,0) - e^{v_n^{[-1]}(t)M^\alpha_\theta}$. Since the commutator $[\pi_2, M^\alpha_\theta] = \pi_2 M^\alpha_\theta - M^\alpha_\theta \pi_2$ vanishes, we have, for every t in $\mathbf{R}$,

$$\begin{aligned}
\|[\pi_2, X^{u_n,\alpha}_{(N)}(t,0)]\| &= \|[\pi_2, e^{v^{[-1]}(t)M^\dagger} + E^{n,\alpha}_{(N)}(t)]\| \\
&= \|[\pi_2, E^{n,\alpha}_{(N)}(t)]\| \leq 2\|E^{n,\alpha}_{(N)}(t)\|.
\end{aligned}$$

Note also that, for every t in $\mathbf{R}$,

$$\begin{aligned}
\|[\pi_2, X^{u_n}_{(N)}(0,t)]\| &= \|X^{u_n}_{(N)}(0,t)[X^{u_n}_{(N)}(t,0), \pi_2]X^{u_n}_{(N)}(0,t)\| \\
&\leq 2\|E^{n,\alpha}_{(N)}(t)\|.
\end{aligned}$$

For every s, t in $\mathbf{R}$,

$$\begin{aligned}
&[\pi_2, X^{u_n}_{(N)}(t,s)] \\
&= \pi_2 X^{u_n}_{(N)}(t,0)X^{u_n}_{(N)}(0,s) - X^{u_n}_{(N)}(t,0)X^{u_n}_{(N)}(0,s)\pi_2 \\
&= X^{u_n}_{(N)}(t,0)[\pi_2, X^{u_n}_{(N)}(0,s)] + [\pi_2, X^{u_n}_{(N)}(t,0)]X^{u_n}_{(N)}(0,s).
\end{aligned}$$

Finally, we get, for every (s,t) in $\mathbf{R}^2$, for every n, N in $\mathbf{N}$.

$$\left\| \left[\pi_2, X^{u_n}_{(N)}(t,s)\right] \right\| \leq 4\|E^{n,\alpha}_{(N)}(t)\|. \quad (18)$$

From (17) and (18), since $\|\pi_2 B(1-\pi_N)\| < \varepsilon/K$,

$$\begin{aligned}
&\|\pi_2 \Upsilon^{\frac{u^*}{n}}(t)\pi_2 - \pi_2 X^{\frac{u^*}{n}}_{(N)}(t,0)\pi_2\| \\
&\quad \leq \varepsilon + 4\|E^{n,\alpha}_{(N)}(t)\| K \|\pi_N B(1-\pi_N)\|. \quad (19)
\end{aligned}$$

From (13), $\sup_{t \leq v_n(K)} \|E^{n,\alpha}_{(N)}(t)\|$ tends to zero as n tends to infinity. For n large enough, for every α in $[0,1]$ and $t \leq v_n(r)$,

$$\|E^{n,\alpha}_{(N)}(t)\| \leq \frac{\varepsilon}{4K\|\pi_N B(1-\pi_N)\|}.$$

Proposition 6 completes the proof of Proposition 1 in the case where (A, B, U, Λ, Φ) satisfies Assumption 2.

C. If A has a mixed spectrum

Assume that $(A, B, \lambda_1, \phi_1, \lambda_2, \phi_2)$ satisfy Assumption 1. From Theorem 2.1, page 525, of [21], for every $\eta > 0$, there exists a skew-adjoint operator A_η such that A_η admits a complete family of eigenvectors (Φ_η) associated with the family of eigenvalues (Λ_η), $A\phi = A_\eta \phi$ for every eigenvector ϕ of A and $\|A - A_\eta\| < \eta$.

For every locally integrable u, we denote with Υ^u_η the propagator of $\frac{d}{dt}\psi = (A_\eta + uB_M)\psi$.

The scheme of the proof is a follows: the result is known (from Section III-B) for the system $(A_\eta, B, \mathbf{R}, \Lambda_\eta, \Phi_\eta)$, which satisfies Assumption 2: we chose $u^* : t \mapsto \cos(\omega t - \theta)$ (this function is the "shape" of the control pulses, it does not depend on η nor ε). For every $\eta, \varepsilon > 0$, θ in $[-\pi, \pi]$ and $r > 0$, there exists an integer n_η and a positive real T_η satisfying, for every α in $[0,1]$,

$$\|\Upsilon^{u^*/n,\alpha,\eta}_{T_\eta,0} - e^{rM^\theta_\alpha}\| < \varepsilon.$$

Notice that, for every t in $\mathbf{R}$, $\|\Upsilon^{u^*/n,\alpha,\eta}_{t,0} - \Upsilon^{u^*/n,\alpha}_{t,0}\| \leq |t|\eta$. The crucial point in the proof of Proposition 1 for systems satisfying Assumption 1 is the existence of a uniform bound on T_η, that depends only on r and ε, and not on η. This follows from (13), where the only variable depending on η is

$$C = \sup_{(j,k)\in\widehat{\Lambda}} \left| \frac{\int_0^T u^*(\tau) e^{\mathrm{i}(\lambda_j - \lambda_k)\tau} \mathrm{d}\tau}{\sin\left(\pi \frac{|\lambda_j - \lambda_k|}{|\lambda_2 - \lambda_1|}\right)} \right|,$$

where $\widehat{\Lambda}$ is the set of all pairs (j,k) in $\{1, \ldots, N\}^2$ such that $b_{jk} \neq 0$ and $\{j,k\} \cap \{1,2\} \neq \emptyset$ and $|\lambda_j - \lambda_k| \notin \mathbf{Z}|\lambda_2 - \lambda_1|$. (Notice that $\|B^{(N)}\|$ is bounded, for every N by $\|B\|$.)

Straightforward computation gives $C \leq 2/d$ where d is the distance of the set $\{2\lambda_1 - \lambda_2, \lambda_1, \lambda_2, 2\lambda_2 - \lambda_1\}$ to the continuous part of the spectrum of iA. This distance is not zero by Assumption 1.6, what concludes the proof of Proposition 1.

IV. EXAMPLE: ROTATION OF A MOLECULE

A. Modeling

The description of the physical system we consider is a toy model inspired by the physical system described in [22]. It has already been thoroughly studied, see for instance [23], [12] or [24]). We consider a polar linear molecule in its ground vibronic state subject to a nonresonant (with respect to the vibronic frequencies) linearly polarized laser field. The control is given by the electric field $E = u(t)(E_1, E_2, E_3)$ depending on time and constant in space. We neglect in this model the polarizability tensor term which corresponds to the field-induced dipole moment (see for instance [25] or [26]).

Let $\mathcal{P}$ be a fixed plane in the space. We are interested in the orientation of the orthogonal projection of a set of molecules in the plane $\mathcal{P}$ (given by *one* angle, in contrary to the orientation of the molecule in the space which is given by two angles). We neglect the interaction between molecules, and consider only the interaction between the molecules and the external field. Our aim is to control the orientation of projection of the molecule in $\mathcal{P}$, whatever the angle of the molecule could be with $\mathcal{P}$.

Up to normalization of physical constants (in particular, in units such that $\hbar = 1$), the dynamics of each molecule is ruled by the equation

$$\mathrm{i}\frac{\partial \psi(\theta,t)}{\partial t} = -\Delta\psi + u_1(t)\cos\theta \sin\varphi \psi(\theta,\varphi,t) \quad (20)$$

where θ is the angular coordinate in $\mathcal{P}$ and φ is the angle of the molecule with $\mathcal{P}$, which is assumed to be constant for the sake of simplicity, Δ is the Laplace–Beltrami operator on the circle $\mathbf{S} = \mathbf{R}/2\pi\mathbf{Z}$, The wavefunction $\psi(\cdot,t)$ evolves in the unit sphere $\mathcal{S}$ of $H = L^2(\mathbf{S}, \mathbf{C})$ endowed with scalar product $\langle f, g\rangle = \int_0^{2\pi} \bar{f}(s)g(s)\mathrm{d}s$.

The operator $A = \mathrm{i}\Delta$ is skew-adjoint in H, with domain $H^2(\mathbf{S}, \mathbf{C})$ and has discrete spectrum. Define $\phi_0 : \theta \mapsto 1/\sqrt{2\pi}$ and, for every k in $\mathbf{N}$, $\phi_{2k-1} : \theta \mapsto \cos(k\theta)/\sqrt{\pi}$ and $\phi_{2k} : \theta \mapsto \sin(k\theta)/\sqrt{\pi}$. The two functions ϕ_{2k+1} and ϕ_{2k} are eigenvectors of A, associated with eigenvalue $-\mathrm{i}k^2$.

The operator $B : \psi \mapsto -\mathrm{i}\cos(\theta)\psi$ is bounded. Straightforward computations show that $|\langle\phi_0, B, \phi_1\rangle| = 1/\sqrt{2}$ and $\langle\phi_j, \phi_k\rangle = 0$ if the parities of j and k are different or if $|j - k| > 2$.

B. Result

Assume that a bunch of molecules is in the state ϕ_0 at $t = 0$. We aim to transfer to the state ϕ_1 all the molecules for which $\varphi > \pi/3$ and to keep all the molecules for which $\varphi < \pi/6$ in the state ϕ_0.

From Proposition 1, applied to $(A, B, 0, \phi_0, 1, \phi_2)$ which satisfies Assumption 1, this is possible, up to the phase and to an arbitrary small error $\varepsilon > 0$.

V. CONCLUSIONS

A. Some comments on the result

While our construction is completely explicit (simple formulas are available for the control laws and come along with precision and time estimates), the convergence toward the target is extremely slow and cannot be used for actual control of real systems. This well-known fact is due to the very poor efficiency of tracking strategies via Lie brackets.

B. Perspectives

The presented results may certainly be improved in many ways. For instance, the author conjectures that it is possible to replace $\hat{\Upsilon}$ in Proposition 1 by a unitary transformation of $\mathcal{L}_N$ with $N > 2$ or to extend the result to systems for which the free Hamiltonian A has a mixed spectrum and the coupling Hamiltonian B is unbounded.

VI. ACKNOWLEDGMENTS

This work has been partially supported by INRIA Nancy-Grand Est, by French Agence National de la Recherche ANR "GCM" program "BLANC-CSD", contract number NT09-504590 and by European Research Council ERC StG 2009 "GeCoMethods", contract number 239748.

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